

# Using Higher Energy X-rays and Spin Detection To Go After Non-magnetic Electron Correlation, Half-metallic Ferromagnetic Behavior and Magnetic Ordering



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**Office of Basic Energy Sciences**

# Outline of Presentation

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- Half-Metallic Ferromagnets and Dilute Magnetic Semiconductors
- Double Polarization and Magnetic Ordering
- Double Polarization and Electron Correlation in Non-Magnetic Systems
- Conclusions

# Instrumentation & Experimental Possibilities

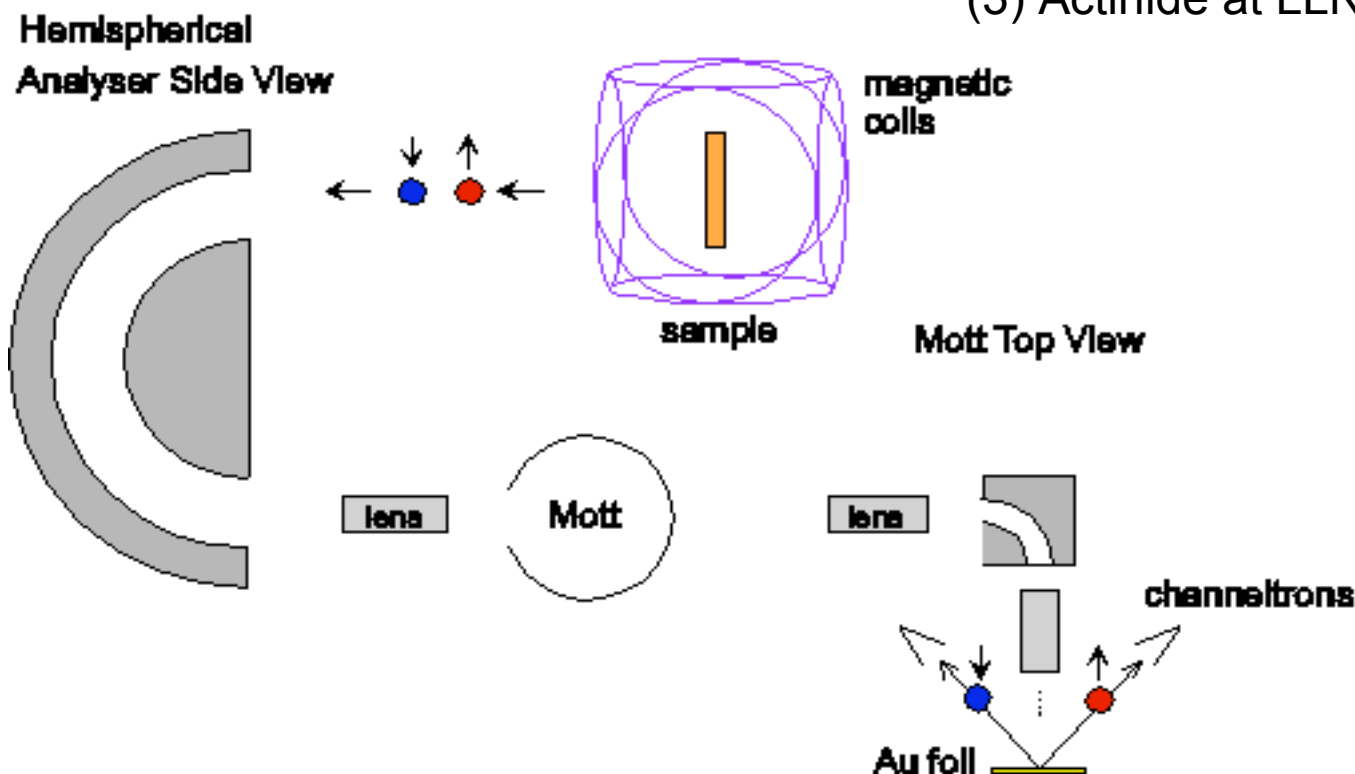


## Spin Spectrometer at the APS

- **Angle-Resolved Photoemission**
- **Spin-Resolved Photoemission**
- **Circular and Linear Dichroism**

Three Spin Resolving Spectrometers:

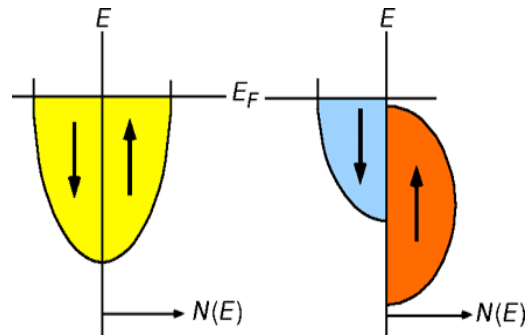
- (1) Spin at BL 4 at the APS
- (2) UMR at BL 4 at the ALS
- (3) Actinide at LLNL



# Half-Metallic FerroMagnets and Dilute Magnetic Semiconductors



What is a half metal?



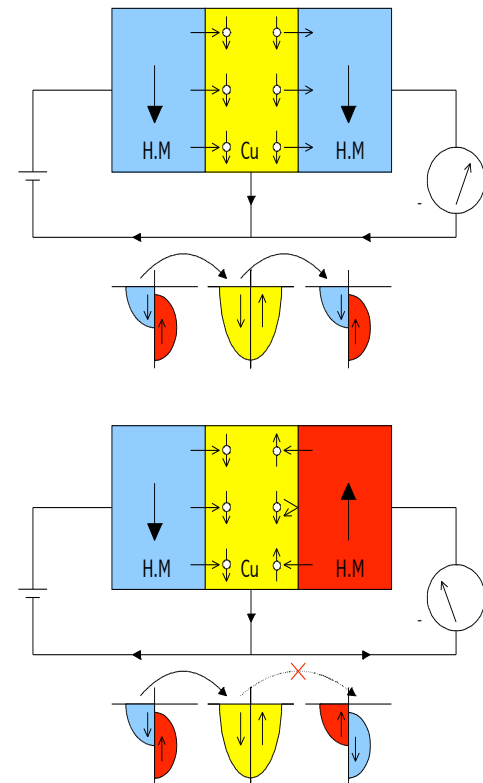
Normal metal

Half metal

- Fermi edge coincides with band gap for one spin population
- Co-existence of metallic behavior for one electron spin and semiconducting for the other
- Electrical properties dominated by single spin
- Unusual transport properties - CMR
- Potential as pure spin source for spin dependant devices

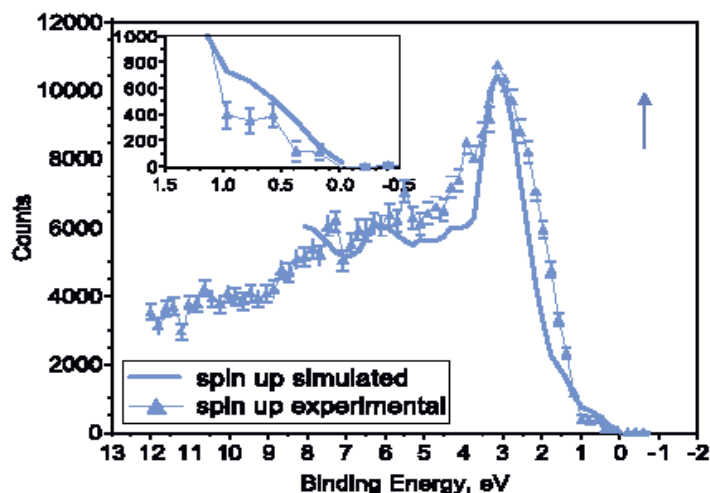
(Prinz, Scientific American P.58, April 1995)

Spin bottleneck magnetoresistive device – spin transistor



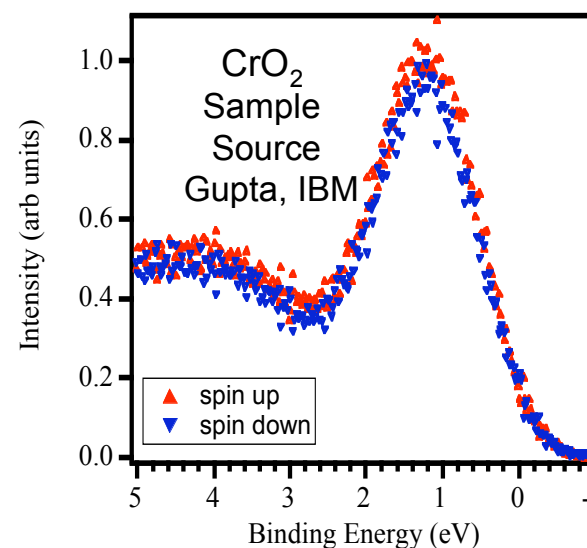
- Magnetic analogue of transistor
- Current in circuit depends on magnetisation of second layer
- Magnetic analogues of other devices also exist - FET
- Can be used to produce non-volatile magnetic analogues of conventional DRAM – MagRAM (IBM)

# Ex situ Prepared Samples: $\text{Fe}_3\text{O}_4$ worked (somewhat) but $\text{CrO}_2$ did not



To the right:  $\text{Fe}_3\text{O}_4$  Sample Source – Schuller *et al*  
At  $h\nu = 160 \text{ eV}$ ,  $\text{Pol} (E_F) = -40\%$

S.A. Morton, G.D. Waddill, S. Kim, I.K. Schuller, S.A. Chambers,  
and J.G. Tobin, *Surface Science Letters* **513**, L451 (2002).



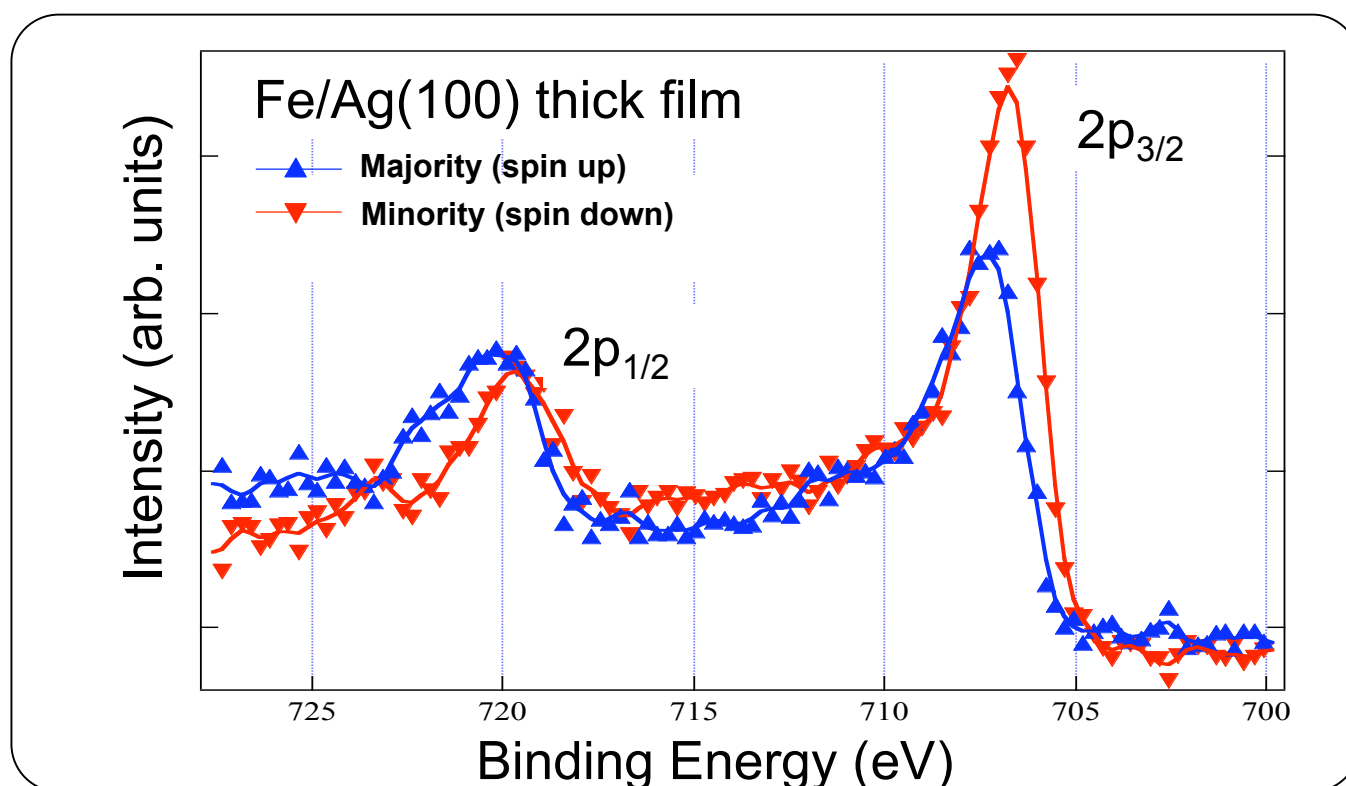
But even the  $\text{Fe}_3\text{O}_4$  result was limited  
by surface effects: we need  
more bulk sensitivity and well behaved  
matrix elements!!! **Higher  $h\nu$ !!!!**

# Spin Polarization In Core Level Photoemission

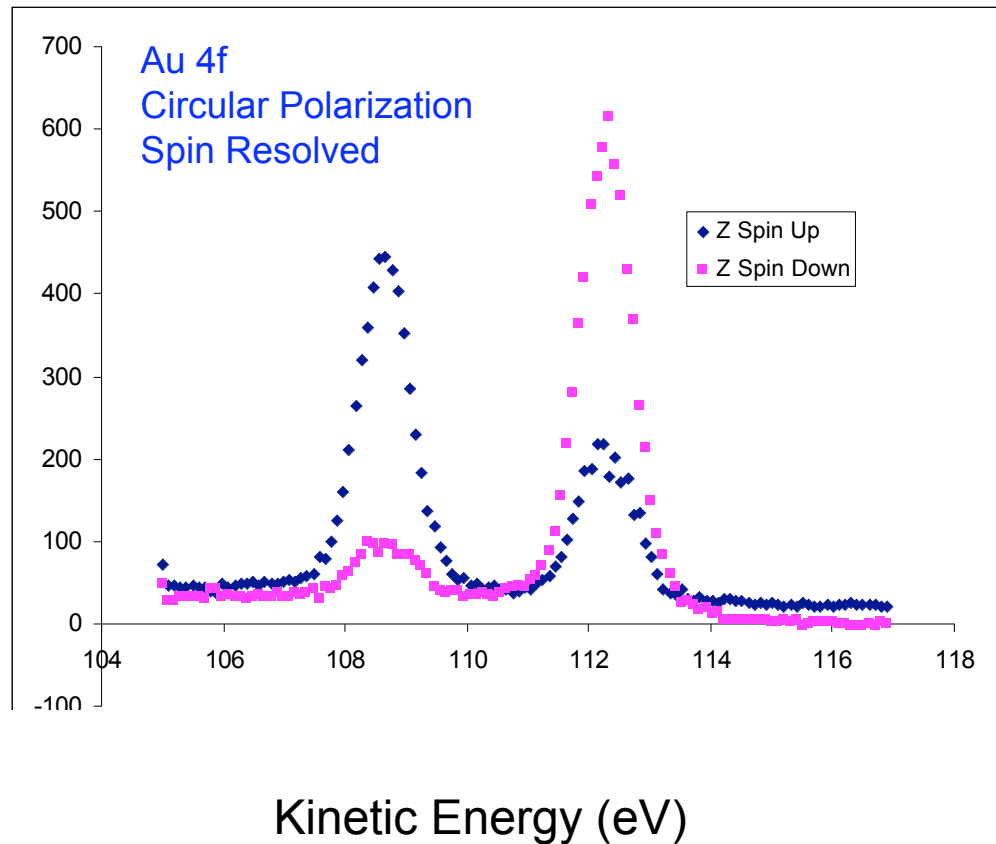
## New Data from BL 4 at the APS



We've seen a strong spin polarization in the Fe 2p's at  $h\nu = 1000$  eV!  
This is a way to probe exchange splittings.



# The Spin Polarization In Core Level Photoemission: A potential source for spin polarized photoelectron diffraction



This data was collected at BL 4 at the ALS, using the UMR Spectrometer and  $h\nu = 200$  eV and circular polarization.

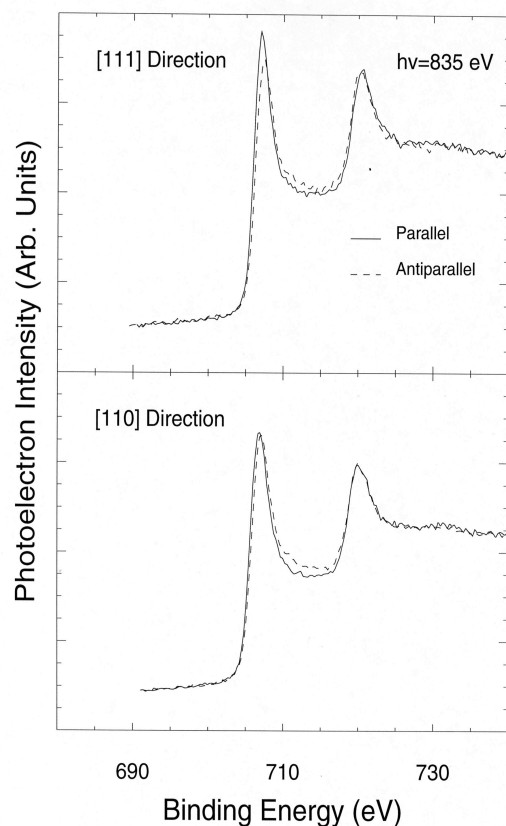
$$B_F(\text{Au}4f_{7/2}) = 84.0 \text{ eV}$$

$$B_F(\text{Au}4f_{5/2}) = 87.6 \text{ eV}$$

# Spin-dependent Photoelectron Diffraction: Circular polarization but no spin resolution



Fe/Cu(001): No spin detection(MCD)  
Small effect: 2% or less

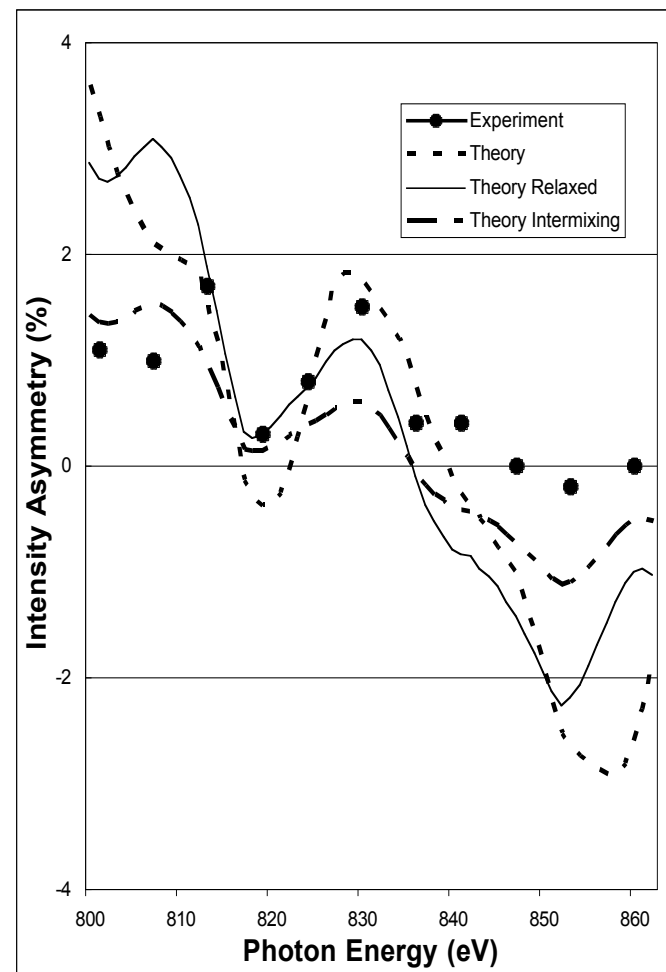


Spin-polarized  
excitation  
using circular  
polarization

Exchange scattering by  
magnetic neighbors

Information on short-  
range spin structure  
around emitter

G.D. Waddill *et al.*,  
PRB **50**, 6774 ('94)

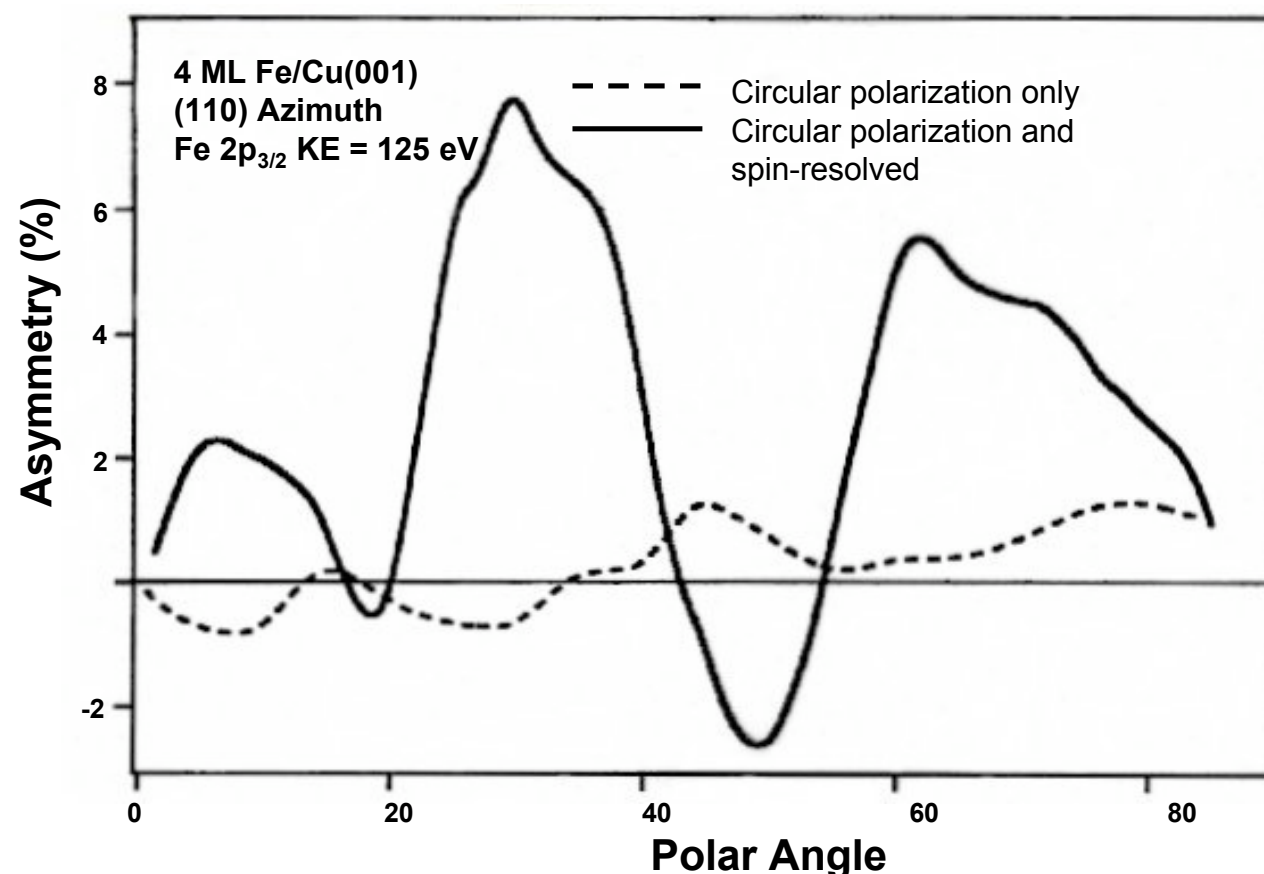




# Spin-Polarized Photoelectron Diffraction: Double Polarization



Theory says that Double-polarized photoelectron diffraction (circularly polarized excitation and true spin detection) will give larger effects.



- With spin-resolution sensitivity to magnetic effects increases 5-10 fold
- Determination of local magnetic structure
- Ultimately imaging magnetic spins with atomic resolution

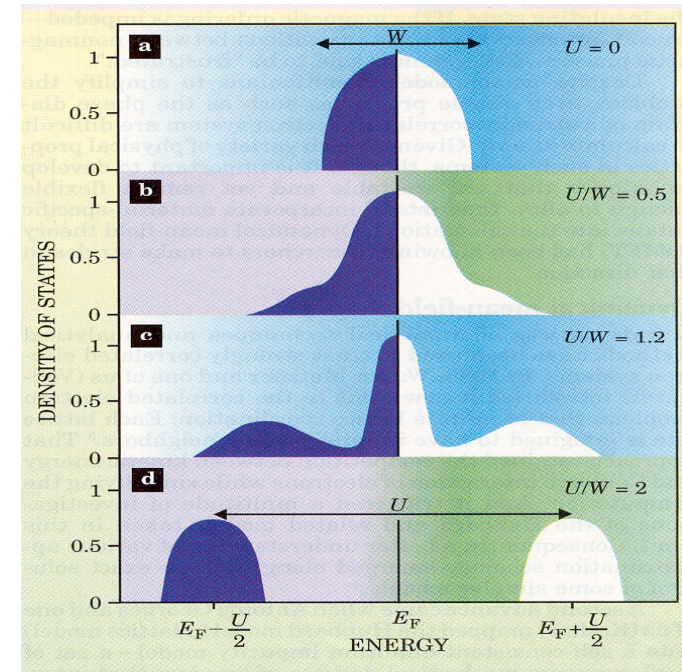
G.D. Waddill *et al.*,  
PRB **50**, 6774 ('94);  
S.Y. Tong *et al.*,  
PRB **54**, 15356 ('96).

# Using the Fano Effect to Probe Electron Correlation in Non-Magnetic Systems



## Electron Correlation in Ce

- Much of the controversy revolves around the interpretation of the Ce photoemission structure in terms of a modified Anderson Impurity Model, first put forth by Gunnarsson and Schoenhammer (PRL and PRB 1983). Here, in this correlated and multi-electronic picture, semi-isolated 4f states ( at a nominal binding energy of 1 eV) are in contact with the bath of spd valence electrons, generating spectral features at the Fermi Level and at a binding energy corresponding to the depth of the bath electron well, about 2 eV below the Fermi Level in the case of Ce.
- To the right, a revisitation to the issue, from Kotliar and Vollhardt, Physics Today, March 2004.



*Illustration of the origin of the quasiparticle ( at the Fermi Level,  $E_F$ ) and the Hubbard Bands (at  $\pm U/2$ , relative to the Fermi Level).  $W$  is the band width and  $U$  is the correlation strength. Case c, third from the top, is the case closest to Ce.*

# The FANO Effect



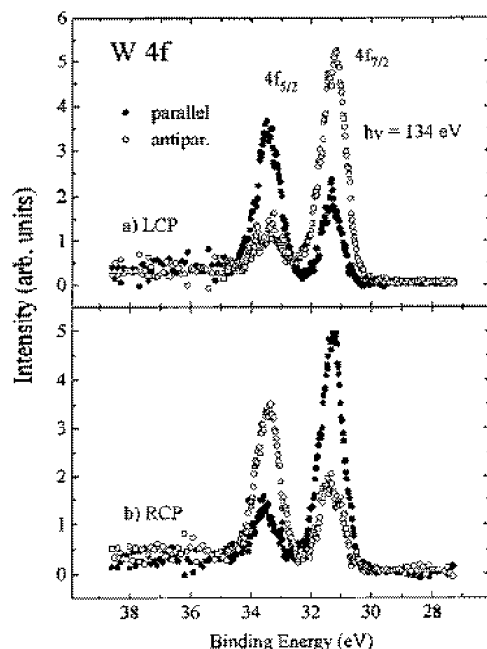
- **The Fano Effect is the observation of spin specific photoelectron emission from the valence bands of a non-magnetic material due to excitation with circularly polarized light.**
- First predicted in 1969 by Fano [U. Fano, Phys. Rev. 178, 131 (1969); 184, 250 (1969)],
- The effect was experimentally confirmed by measuring the polarization of alkali vapor beams using detection of ions [M.S. Lubell and W. Raith, Phys. Rev. Lett. 23, 211 (1969); J. Kessler and J. Lorenz, Phys. Rev. Lett. 24, 87 (1970); G. Baum, M.S. Lubell, and W. Raith, Phys. Rev. Lett. 25, 267 (1970)]
- The effect was more directly confirmed by measuring the spin polarization of photoelectron emission shortly thereafter [U. Heinzmann, J. Kessler, and J. Lorenz, Phys. Rev. Lett. 25, 1325 (1970)].

# Circularly polarized x-rays and PES: the Fano effect in Core Levels of NON-MAG materials

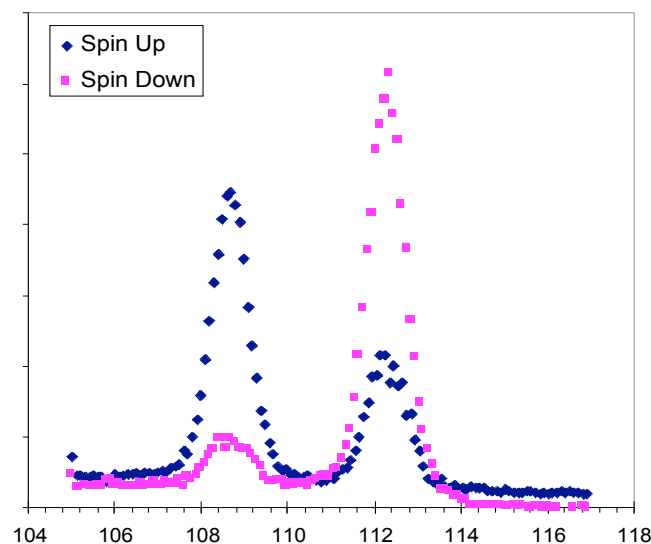


## Double Polarization Photoelectron Spectroscopy: W 4f and Au 4f

W 4f with Circular Polarization:  
K. Starke et al, Phys. Rev. B 53,  
10544 (1996).



Au 4f with Circular Polarization:  
The UMR Spectrometer on BL 4  
EPU at the ALS



# Fano Effect in nonmagnetic Ce



- Polycrystalline, non-magnetic Ce/W(110)
- Circularly polarized x-rays from the EPU at the ALS (Berkeley)... $h\nu = 127$  eV, on resonance
- Strong effect along x, no effect along y, as expected.
- The peak at  $KE = 121$  eV ( $BE = 2$  eV) shows the same simple relationship as the Au 4f's but the peak at the Fermi Level has a different dependence...
- The peak at  $BE = 2$  eV ( $KE = 121$  eV) appears to be broadened: this could be due to multiple phases, surface versus bulk effects or strain in the overlayer.

**Following the lead of Mo et al (PRL 2003), we'd like to extend these measurements to higher energies.**

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- **To the right are some of our spectra of Ce thin films. These spectra were collected using Beamline 4 at the APS.**
- **The intensities of the quasiparticle peak (near  $E_F$ ) and lower Hubbard band ( $B_F = -2$  eV) seem to have a different  $h\nu$  dependence.**

# More high hv spectra of Ce

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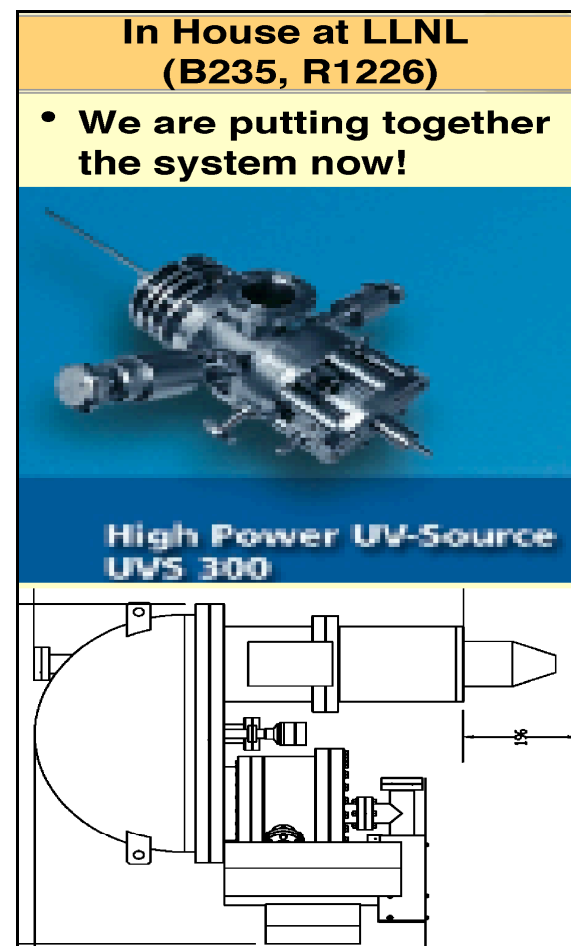
- A more detailed look between 700 and 800 eV
- These are spin integrated spectra: we are aiming to do spin resolved when the new refocusing mirror is installed.

# Ultimately, we'd like to extend these measurements to Pu



**Our new model  
predicts a Fano  
Effect for Pu!**

- We can explain conventional PES results for Pu with our New Paradigm Model
- Top: Bulk Pu,  $h\nu = 125\text{eV}$ , J.G. Tobin et al, PRB
- Middle panel: 1ML Pu/Mg,  $h\nu = 40.8\text{ eV}$ , Gouder et al, EuroPhysLett
- Bottom panel: Our New Theory: Individual  $5f^{5/2}$  states are resolved and shown as colors. The sum is shown as black.





# Conclusions: the advantages of higher energies

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- **Half-Metallic Ferromagnets and Dilute Magnetic Semiconductors**
  - **Eliminate surface effects to get a bulk polarization**
- **Double Polarization and Magnetic Ordering**
  - **Increase the effect to 10% - 20%...Image 3d magnetic structure with 2p electrons**
- **Double Polarization and Electron Correlation in Non-Magnetic Systems**
  - **New probe of electron correlation in nonmagnetic systems, with bulk sensitivity**